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NOVEL LIQUID CRYSTALLINE STRUCTURE FOR NONLINEAR OPTICS

I. C. Khoo

Department of Electrical Engineering
121 Electrical Engineering East
The Pennsylvania State University
University Park, PA 16802

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PHILLIPS LABORATORY
Lasers and Imaging Directorate
AIR FORCE MATERIEL COMMAND
KIRTLAND AIR FORCE BASE, NM 87117-5776

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This report has been approved for publication.

Christopher M. Clayton

CHRISTOPHER M. CLAYTON
Project Manager

FOR THE COMMANDER

Marc R. Hallada

MARC R. HALLADA, LtCol, USAF
Chief, Laser Systems Division

Robert A. Duryea

ROBERT A. DURYEA, Col, USAF
Director, Lasers and Imaging Directorate

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14. Abstract This effort consisted of both theoretical and experimental studies of several novel nonlinear optical phenomena in liquid crystal fiber structure and thin films, exploring their potentials for nonlinear-, electro- and adaptive-optical applications. Continuous and pulsed lasers with pulse durations spanning the nanosecond (ns) to picosecond (ps) regime were employed to study these processes in both the ordered (nematic) and the isotropic (liquid) phases of liquid crystals. The projects completed include: characterization of optical nonlinearities of these liquid crystalline materials, studies of coherent beam amplification, phase conjugation effects, and quantitative characterization of their responses with ns and ps laser pulses. The nonlinear fiber arrays were shown to be capable of good quality image transmission while serving as an optical limiter for short intense pulses throughout the visible spectrum. This research program has also resulted in the discovery of orientational photorefractive effects which will be relevant to current research and development in adaptive optics and holographic storage systems.

15. Subject Terms nonlinear optics, liquid crystals, coherent beam amplification, phase conjugation, photorefractive effects

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1.0. PROJECT SUMMARY, METHODS AND PROCEDURES

This effort consisted of both theoretical and experimental studies of several novel nonlinear optical phenomena in liquid crystal fiber structure and thin films and explored their potential and feasibilities for nonlinear-, electro- and adaptive-optical applications. A variety of lasers (cw, nanosecond- and picosecond-pulsed system) in the visible-near infrared regime were employed to study these processes in both the ordered (nematic) and the isotropic (liquid) phase of liquid crystals. The projects completed are broadly grouped under the two following categories.

a. Liquid crystals-dye and other dopant molecules optical nonlinearities

The contractor conducted further studies of coherent beam amplification and phase conjugation effects in nematic liquid crystals doped with the appropriate dyes; the enhanced thermal as well as orientational nonlinearities of these were investigated using visible and near infrared millisecond or shorter laser pulses. The broad spectral bandwidth applicability of these phenomena and liquid crystals as well as the much faster response of liquid crystal in comparison to other phase conjugation media (e.g., photorefractive crystals) were quantitatively documented.

Using lasers spanning a wide time scale--from cw to picosecond--and spectral regime--visible to infrared--the contractor quantitatively characterized the mechanism(s) responsible for enhanced orientation response of dye-doped or absorbing liquid crystals to polarized lasers. In particular, the differences and similarities between systems with positive "dye-induced torque" (liquid crystals with AQ_1 and AQ_2) and negative "dye-induced torque" (liquid crystals with dichroic dyes) were documented in temporal and polarization discriminating studies.

Conditions whereby these nonlinear optical responses could be optimized were determined; the efficiency, limitations, and response times in the ordered (nematic) and the disordered (isotropic) phase near T_c were measured and theoretical formalisms for their quantitative descriptions were developed. The contractor obtained detailed understandings of these dye-doped liquid crystals, pure liquid crystals and liquid crystal mixtures. Their nonlinear optical responses are crucial in optical wave mixings, self-starting phase conjugation effects and holographic grating formation processes.

b. Liquid crystalline optical fibers

The contractor further developed and refined the techniques for fabricating liquid

crystalline optical fiber structures using liquid crystals in the aligned phase (nematics and smectics) and the isotropic (liquid) phase. The principal goal was to ascertain the choice of configuration/geometry and liquid crystalline materials that would yield low optical propagation loss and high nonlinearities. To achieve that, the contractor experimented with pure liquid crystals, liquid-crystal mixtures, and dye-doped liquid crystals in miniature-cored fibers of various core diameters and length. The dimensions of the fibers were dictated by the vastly different magnitudes of the scattering losses and nonlinearities in these mesophases.

Following a quantitative characterization of the liquid crystal nonlinear optical fiber, orientation scattering experiments were conducted with ns and ps Nd:Yag laser pulses (at 1.06 fundamental and 0.53 second harmonics). Greatly reduced threshold for stimulated scatterings and other nonlinear propagation effects were observed as a result of the longer interaction length as well as more nonlinear dye-liquid crystalline materials used. By varying the interaction length, core diameter (which changes the single- or multi-mode guided wave characteristics), optical intensity, dopant concentration, temperatures near T_c, phases, etc., variant of stimulated scattering effects were sought. Optical phase conjugations, pulse compression and stretching, and intense phase modulation effects, and intensity dependent propagation modes were but a few examples of some of the interesting possibilities investigated by the contractor.

2.0. STATEMENT OF TASKS

This contractor has performed experimental and theoretical studies aimed at exploring and exploiting nonlinear optical phenomena occurring in dye-doped liquid crystals, and liquid crystal optical fibers for novel coherent optical devices or applications. CW and pulsed lasers covering the visible through the near infrared regime were employed to study the material and structures for various time regimes, intensity levels and geometrical/optical configurations.

2.1. Basic Program

The contractor experimentally investigated various techniques at fabricating optical quality liquid crystal fibers, and various dye-doped liquid crystalline systems. The linear and nonlinear optical properties of these materials/structures were quantitatively documented. The contractor performed further studies of recently observed novel optical wave mixing and stimulated scattering effects, and all-optical switching effects with short pulses.

2.2. Phase 1

The contractor employed techniques developed in his laboratory to fabricate liquid crystalline fibers of various core dimensions and length, in conjunction with several liquid crystalline materials that have been previously shown to exhibit large optical nonlinearity and/or novel optical responses. A variety of dichroic or azo-dyes were introduced into these liquid crystals to improve the performance of these materials or increase their nonlinearities. The parameters measure/documented include propagation loss, nonlinearity, response times, power/intensity thresholds for nonlinear effects, laser pulse width required, and a variety of dye/liquid crystal parameters.

2.3. Phase 2

The contractor performed experiments to elucidate stimulated backward scattering and phase conjugation, and forward guided/stimulated scattering effects in liquid crystalline fibers under submillisecond and nanosecond laser pulse excitations. The contractor performed experiments to qualitatively characterize the self-pumped optical phase conjugation effects, and the process of laser writing of storable gratings in dye-doped liquid crystal films.

2.4 Phase 3

The contractor performed experiments to further establish these novel materials/phenomena; in particular, complete theories, with experimental substantiations, were developed to describe stimulated scatterings and phase conjugation effects, erasable and reconfigurable optical grating or holographic element formations.

3.0 RESULTS

The 3-year program just concluded has resulted in several first time observations and discoveries in liquid crystalline films and fiber structures. The main work performed during this period was: studies of nonlinear propagation in C_{60}-doped liquid crystal fiber, transient and permanent grating/holographic image formation in C_{60}-doped nematic liquid crystal film, detailed theories of self-starting optical phase conjugation in nematic films with thermal and stimulated orientational scattering effects, fabrication of liquid-crystal cored fibers and fiber arrays, all-optical switching/limiting and nonlinear propagation, stimulated backscatterings with nano- and subnano-second laser pulses in these fiber structures.

3.1 Nonlinear Optical Effects in Liquid Crystalline Fibers and Fiber Array

Liquid crystals are composed of large and complex molecules and possess unusually large optical nonlinearities with unique characteristics. Consequently, several fundamentally interesting and practically important nonlinear optical phenomena have been observed in bulk nematic liquid crystal thin films [5.1]. These include temporal and spatial self phase modulations, optical phase conjugation, image/beam amplifications and modulations, parametric interactions polarization switching, bistabilities and instabilities, and pulse propagation.

These novel physical responses of liquid crystals, and the optical phenomena, will take on brand new dimensions and assume even more unusual and novel characteristics if the liquid crystals are fabricated into guided wave structures (fiber or planar waveguide). Much enhanced efficiency, miniaturization and compatibility with other integrated optical components are but a few of the obvious advantages. Since the optical nonlinearities (e.g. the picosecond-electronic contribution) of liquid crystals is more than 10^6 times that of the usual glass materials used for fabricating optical fibers, many navel nonlinear optical phenomena have been shown to take place with much greater efficiency in liquid crystalline fibers. Processes that used to require kilometers of fibers, such as pulse stretching and compressions, nonlinear soliton interactions and propagation, etc., can now be made to occur within a few millimeters of liquid crystalline fibers, i.e., a dramatic miniaturization in the physical dimension and reduction in the power requirement. Also, the threshold for a variety of coherent all-optical switching processes were greatly lowered, while the signal levels were greatly enhanced.

Nonlinear fibers and fiber arrays were fabricated by filling micro-capillaries and capillary arrays with liquid crystalline materials; solid state liquid crystalline fiber structures were made with special LC-polymer mixture as the core filling material. These arrays were inserted at the image plane of an optical system to test their usefulness for optical sensor protection application, and electro-optically controllable image transmission and modulation devices [5.2].

The contractor also introduced laser-dyes, dichroic dyes and Fullerene C_{60} as dopants in the liquid crystalline cores in these fiber structures and observed pronounced reduction in the threshold requirement for optical switching and limiting action with nanosecond laser

pulses. The fundamental nonlinear mechanisms, as probed by 100 ps and 20 ns laser pulses [5.3-5.5] were determined to be thermal-density effects following laser absorption in dopant dye molecules, nonlinear [two- and multi-photon] absorption, reverse saturable absorption in C_{60}. A quantitative theoretical description of the nonlinear propagation in such fibers and comparison with experimental observation was developed [5.6].

The contractor conducted characterization of the linear and nonlinear properties of these arrays. Results showed that they functioned as high quality image transmitting faceplate as well as low threshold all-optical switching/limiting elements [5.7]. The contractor obtained for the first time nonlinear optical switching of nanosecond laser pulses through such fiber array; the switching threshold obtained in an unoptimized sample was already very low [~ μJ], and ranks among the lowest of all existing materials under consideration for limiting application. In the last period, the contractor has also obtained for the first time nonlinear propagation and optical switching/limiting of picosecond laser pulses through such fibers, and even lower switching thresholds [<0.1 μJ]

A series of studies on other liquid crystals and dopants [besides dyes and C_{60}] as the nonlinear core materials has also been conducted, with the goals of improving the nonlinear switching thresholds of these fiber structures. Preliminary studies have identified two particularly nonlinear liquid crystals [a alkyl tolane] and a mixture [ILC doped with an absorbing LC] that yielded very low switching threshold. Currently, the contractor is investigating their spectral, two-photon absorption efficiency, and other linear and nonlinear optical properties.

3.2 Nonlinear Optical Effects in Dye-, Fullerene-Doped Liquid Crystal Films

Self-starting optical phase conjugations [SSOPC] in nematic films, originating from thermal and orientational wave mixing or stimulated scatterings mechanisms, have been theoretically and experimentally investigated in details. The contractor has reported first-time observation of SSOPC in pure non-absorbing films [stimulated orientational effect][5.8] and dye-doped film [thermal grating] [5.9], and has developed detailed theoretical formalisms that show good agreement with the experimental results. These theories and experimental results are being organized into a paper for publication in J. Opt. Soc. Am. B .

During the course of this program, the contractor has discovered a new nonlinear electro-optical effect in pure, dye- or fullerene C_{60} doped nematic liquid crystal films, namely,

orientational photorefractive effects. The effect required very low applied dc field [~ 1 volt in a 100 μm thick film] [5.10,5.11] and optical field [~ 1 m Watt] unfocused cw laser beam. The contractor has performed photoionization and photoconduction experiments , and developed theories for the space charge fields and nonlinear electro-optical induced refractive index changes in these fullerene- and dye-doped nematic liquid crystal films. The theories showed good agreement with experimental studies of the dynamics and geometrical discrepancies of the effects in the transient as well as storage modes [5.12].

Studies [5.11] have shown that in these materials, the electro-optically induced grating could persist for a long period of time [several months by now]. These persistent gratings could be electrically turned on/off, or modulated with ac/dc field, and response in a very interesting manner as the frequency of the ac field is varied from a few Hz to MHZ. Furthermore, preliminary studies have shown that persistent grating with grating constant as low as 10 microns can be generated, that will still produce fairly good diffraction efficiency [a few percents].

The contractor investigated various other material and optical aspects of photorefractive effects in dye- and fullerene-doped nematic films with different Argon laser lines [visible - UV] and has also begun studies of nematic films doped with IR absorbing dyes to investigate the possibility of observing photorefractivity in the 1.3-1.5 μm regime. A few IR dyes have been identified, and they are being tested for their photoconductivities.

3.3 Graduate Student Training

An important accomplishment of this research program is the training of graduate students, resulting in three Ph.D. (Dr. Y. Liang, Dr. P. LoPresti and Dr. H. Li) and one Master Theses (Mr. B. Yarnell). These students have been partially supported by this program at various stages of their graduate studies.

4.0 CONCLUSIONS AND RECOMMENDATIONS

Besides completing all the proposed major tasks, this research program has also resulted in several new important findings, discoveries, and opened up new avenues for future research. In particular, the photorefractive effects observed in dye- or C_{60}-doped nematic films will be relevant to current research and development in holographic storage. On the other hand, the nonlinear fiber and fiber arrays could be further developed for applications in sensor protection systems, as optical

limiter for short pulses.There are many competitive advantages (low threshold, large dynamic range, fast [ns or sub-ns], broadband) that liquid crystal fiber and fiber arrays could offer in comparison with other materials currently being investigated for sensor protection or laser hardening applications.

Currently several classes of materials are actively being investigated for their capabilties of recording real time or permanent holographic gratings. Examples are inorganic and plymeric photorefracrtve crystals, bateriorhodopsin, semiconductors and doped glasses, as well as liquid crystals. These materials possess various advantages but also limitations. The main limitations are the working spectral range. To date, photorefractive crystals work in the range from visible to around 1μm. By using IR absorbing dyes in nematic film, it is possible to extend the working spectral range to the communication channel (1.3-1.5 μm). Also the effects discovered by the contractor requires much lower dc field than can be drived from batteries, in comparison with polymeric film and inorganic photorefractive crystals, which require very high poling dc field to work. These and other useful aspects of photorefractive effects in dye-doped or C_{60}-doped nematic film were fully discussed in a proposal submitted to Air Force Phillips Laboratory with a project start date of around March 1996.

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